MM 12: Materials for the Storage and Conversion of Energy III / Functional Materials I

Time: Tuesday 10:15–12:45 Location: SCH/A216

MM 12.1 Tue 10:15 SCH/A216

Harmonising Symmetry and Spin: Unveiling Giant Rashba Splitting and Diverse Topological Phases in Composite Quantum Materials — •Arijeet Sarangi, Rajib Sarkar, and Sudip Chakraborty — Harish-Chandra Research Institute, Chhatnag Road, Jhunsi, Prayagraj - 211019, India

We present a systematic theoretical study of ABX-type composite quantum materials crystallizing in non-centrosymmetric hexagonal structures, carried out through high-throughput DFT calculations. Approximately 50 candidate systems were screened to explore the interplay between structural symmetry breaking, strong SOC and emergent electronic properties. Our investigation reveals a striking coexistence of Rashba spin splitting and topologically nontrivial band topology across this materials family, with Rashba coefficients and band inversions strongly dependent on atomic ordering and chemical composition. To complement the band-structure analysis, we perform Berry phase calculations, which confirm the topological character of selected states and identify Berry curvature hotspots near avoided crossings. Building upon this, we evaluate the spin Hall conductivity and orbital Hall conductivity for representative compounds, demonstrating substantial intrinsic values comparable to those in established topological semimetals. These findings point to a strong potential for dissipationless spin and orbital current generation, key ingredients for next-generation spintronic and orbitronic devices. These results establish a promising design space for multifunctional applications, bridging spintronics and topological electronics.

MM 12.2 Tue 10:30 SCH/A216

TiO_2 meets water: Band alignment and spectral properties for solar-driven catalysis using Koopmans spectral functionals — •Marija Stojkovic[1], Edward Linscott[2,3], and Nicola Marzari[1,2,3] — [1]Theory and Simulations of Materials (THEOS), École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland — [2]Center for Scientific Computing, Theory and Data, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland — [3]National Centre for Computational Design and Discovery of Novel Materials (MARVEL), Paul Scherrer Institute, 5232 Villigen PSI, Switzerland

Photocatalytic water splitting has drawn considerable attention for renewable energy production. Thanks to its favorable band gap and band-edge positions, titanium dioxide has remained one of the most promising materials since its first reported photocatalytic water splitting. However, conventional computational approaches often struggle to accurately predict band gaps and band alignments. Here, we showcase the capabilities of Koopmans spectral functionals for evaluating photocatalysts* properties, showing how these can accurately predict band structures and level alignments for rutile, anatase, and brookite TiO_2. Furthermore, we investigate the impact of explicit water on band alignment at TiO_2-water interface by combining advanced molecular dynamics methods with electronic-structure Koopmans calculations.

MM 12.3 Tue 10:45 SCH/A216

Theoretical Calculation of Finite-Temperature X-Ray Absorption Fine Structure: Application to Sodium K-Edge in NaCl — Philipp Hönicke[1,2], Yves Kayser[3], and •Pouya Partovi-Azar[4] — [1]Helmholtz-Zentrum Berlin (HZB), Hahn-Meitner-Platz 1, 14109 Berlin, Germany — [2]Physikalisch-Technische Bundesanstalt (PTB), Abbestr. 2-12, 10587 Berlin, Germany — [3]Max Planck Institute for Chemical Energy Conversion, Stiftstr. 34-36, 45470 Mülheim an der Ruhr, Germany — [4]Institute of Chemistry, Martin Luther University Halle-Wittenberg, Von-Danckelmann-Platz 4, 06120 Halle (Saale), Germany

A comprehensive computational framework for calculating the full X-ray absorption fine structure (XAFS) through quantum-chemical simulations is presented. The near-edge region is accurately captured using time-dependent density-functional perturbation theory applied to core excitations, while *ab initio* molecular dynamics provides sampling of core-excitation energies and interatomic distance distributions for interpreting extended X-ray absorption fine structure (EXAFS) features. Owing to the efficiency of the approach, the total spectrum can be decomposed into contributions from bulk, defective, and surface environments, which commonly coexist in experimental systems. The methodology is demonstrated for sodium at the Na K-edge in NaCl, where the predicted spectra show good agreement with experimental measurements on thin film samples. This strategy offers a practical route to generating chemically specific XAFS cross-section data for elements that remain challenging to characterize experimentally.

MM 12.4 Tue 11:00 SCH/A216

Spin-Charge Coupling at Mixed Fe/Cr Nodes Governs Optical Gaps in MIL-100 Photocatalysts — •Sabuhi Badalov[1], Patrick Länger[2], Jürgen Senker[2], and Harald Oberhofer[1] — [1]Chair for Theoretical Physics VII and Bavarian Center for Battery Technologies, University of Bayreuth — [2]Chair for Inorganic Chemistry III and Northern Bavarian NMR Centre, University of Bayreuth

With a view on photocatalytic CO_2 reduction, we investigate how mixed metal Fe/Cr nodes in MIL-100 metal organic frameworks (MOFs) influence the MOFs electronic and optical response. Using hybrid level density functional theory (DFT) on models of Fe_3O, Fe_2CrO, $FeCr_2O$ and Cr_3O, we map the evolution of spin-resolved edge levels and Kohn-Sham holes during the stepwise substitution of Fe with Cr. Time-dependent DFT calculations reveal dense $O(2p)\rightarrow M(3d)$ ligand-metal charge transfer (LMCT) bands, appearing between approximately 3.5 and 4.5 eV. From these we derive composition-dependent optical band gaps and λ_{max} values. The calculated trends, modest red/blue shifts of the first bright excitation and systematic hardening of the LMCT manifold with increasing Cr content, quantitatively agree with the experimentally obtained gaps. Our combined theoretical/experimental approach provides a deep microscopic insight into the influence of the Fe/Cr mixed metal centers on spin topology, band edges, and optical absorption. Ultimately, our work points the way towards a rational design of MIL-100 for maximal CO_2 conversion.

15 min. break

MM 12.5 Tue 11:30 SCH/A216

Beyond Ion Dynamics: Efficient Charge Transport Simulations including Electrons at Battery Scales — Matteo Rinaldi, Karsten Reuter, and •Christian Carbogno — Fritz-Haber-Institut der MPG, Berlin

Small polarons, i.e., atomically localized excess charges, are fundamental for charge transport in energy materials. A quantitative modeling of their dynamics is challenging, though, since it typically requires not only long time and length scales, but also an account of electronic degrees of freedom at an *ab initio* level. In this work, we overcome this hurdle via a machine-learning interatomic potential that explicitly accounts for small polarons, which are incorporated as semi-classical degrees of freedom that adiabatically follow the nuclei. Using HSE06 calculations [1] as training data, we validate the approach for $BiVO_4$ and further demonstrate its power for lithium titanium oxide (LTO), a prototypical anode material featuring polarons [2]. By running accelerated sampling and large-scale molecular dynamics simulations, we clarify the mechanisms that drive ionic and polaronic transport as well as their coupling in LTO. This reveals that polarons do not merely serve as spectators, but thermodynamically adapt to the much slower ionic motion. This results in an increase of ionic conductivity, in line with experimental measurements. For the first time, this theoretically corroborates the occurrence of a correlated polaron-ion dynamics with profound implications for the design of energy materials.

[1] S. Kokott *et al.*, J. Chem. Phys. **161**, 024112 (2024).
[2] M. Kick *et al.*, ACS Appl. Energy Mater. **4**, 8583 (2021).

MM 12.6 Tue 11:45 SCH/A216

Ion Transport in Mixed-Halide Lithium Argyrodites from Machine Learning Potentials — •Yufeng Xu, Takeru Miyagawa, Waldemar Kaiser, and David A. Egger — Physics Department, TUM School of Natural Sciences, Technical University of Munich, 85748 Garching, Germany

Lithium argyrodites Li_6PS_5X (X=Cl, Br, I) are structurally and chemically complex solid electrolytes. Their ionic conductivity is strongly affected by static forms of disorder, including sulfur-halide anti-site defects [1] and compositional disorder introduced through halide mixing [2]. In this study, we use the MACE message-passing architecture [3] together with machine-learning molecular dynamics to examine ion transport mechanisms in both pure and mixed-halide Li_6PS_5X. We analyze how static and dynamic disorder in mixed-halide systems shapes Li ion diffusion, and how these changes in ion conduction are reflected in the vibrational spectra of the argyrodites. References: [1] B.J. Morgan, Chem. Mater., 2021, 33, 6, 2004-2018. [2] S.V. Patel et al. Chem. Mater., 2021, 33, 4, 1435-1443. [3] I. Batatia et al. J. Chem. Phys., 2025, 163, 184110.

MM 12.7 Tue 12:00 SCH/A216

Positron Annihilation Studies of Nuclear and Highly Oriented Pyrolytic Graphite — •Jagoda Urban-Klaehn, Arvin Cunningham, and William Windes — Idaho National Laboratory, 1955 Fremont Ave, Idaho Falls, ID 83415 USA

Graphite is a life-limiting component in nuclear reactor design thus its microstructure characterization and the effects of extreme conditions are of great importance.

We examined nuclear graphite materials of different origin, porosity and grain sizes by use of Positron Annihilation Lifetime Spectroscopy (PALS).

We determined that each type of graphite has its specific signature with trapping lifetimes in the range: 360-410 ps, and the most important factor was the grain size. Graphite with the coarse grain showed less positron trapping compared to fine grain which agrees with literature.

Graphite that was heat-treated (2300 C) showed a significant increase in positron trapping thus changes in the microstructure. PALS studies on Highly Oriented Pyrolytic Graphite (HOPG) polycrystalline specimens showed less trapping due to its polycrystalline nature. We are planning to use ELBE slow positron beam at different temperatures to study isotropic graphite defects formation enthalpy as well as crystalline graphite*s depth profiling.

MM 12.8 Tue 12:15 SCH/A216

Positron annihilation spectroscopy for atomic defects in functional materials — •Maciej Oskar Liedke, Eric Hirschmann, Jyoti Verma, Sebastian Klug, and Andreas Wagner — Institute of Radiation Physics, Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstraße 400, 01328, Germany

Positron annihilation spectroscopy (PAS) is a highly effective method for the characterization of point defects and their agglomerates in functional bulk materials and thin films. The method provides access to defect concentration in the range of 0.01-100 ppm, which is common for most material classes, including metals, semi-, and superconductors. The utilization of the radiation source ELBE at HZDR in Dresden, which delivers a pulsed positron beam, enables depth profiling by varying the kinetic energy of positrons. The integration of PAS techniques with density functional theory calculations facilitates understanding of defect types and local chemistry at the defected crystal sites. In this contribution, we will present few highlights from positron studies and discuss the role of defects in wide-bandgap semiconductors, such as Ga2O3, in addition to superconducting Nb-based coatings, and high-entropy alloys intended for fusion applications. The presentation will emphasize the control over transitions between different polymorphs of Ga2O3 driven by ion irradiation and resulting defect microstructure. Furthermore, we will demonstrate the correlations between deposition parameters and defect landscapes in Nb alloys. We will conclude discussing healing processes in CrFeMnNi subjected to Fe-ion irradiation, which is analogous to neutron radiation in nuclear systems.

MM 12.9 Tue 12:30 SCH/A216

Operando X-ray Absorption Spectroscopy studies of the X-ray exposed active and passive fiber preforms and their recovery properties upon UV-laser light — •Ali KARATUTLU[1,2], Esra KENDIR TEKGÜL[1], Iremnur DURU[1,2], Timuçin Emre TABARU[2], Latif Ullah KHAN[3], Messaoud HARFOUCHE[3], and Bülend ORTAÇ[1] — [1]Bilkent University, Institute of Materials Science Nanotechnology and National Nanotechnology Research Center (UNAM), Ankara, 06800 Turkey — [2]Sivas University of Science and Technology, Department of Engineering Basic Sciences, Sivas, 58100 Turkey — [3]Synchrotron-light for Experimental Science and Applications in the Middle East (SESAME), Allan, 19252, Jordan

In the development of active fiber preforms and active fibers containing rare-earth (RE) ions such as Yb+3 ions for 1-micron laser applications, the determination of the local environment around the Yb+3 ions is of paramount importance for their high optical-optical efficiency. In the long-term operation or when exposed to the gamma or X-ray exposures, the unwanted structural units are usually associated with the defects called color centers. This study demonstrates the formation of photodarkening upon gamma and X-ray exposures and operando X-ray absorption spectroscopy to determine the structural change around Yb+3 ions or Ge+4 ions with different doping levels before and after the irradiation of X-rays.